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1 **Nucleation delay in water-saturated rhyolite during decompression in shallow volcanic systems**  
2 **and its implications for ascent dynamics**

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8

9 **Abstract**

10 We developed a model of feldspar nucleation delay during decompression in water-saturated rhyolite  
11 based on classical nucleation theory (CNT). With the thermodynamic and kinetic parameters proposed  
12 in this study, the model agrees with the experimental results of this study to within a factor of two. To  
13 account for the variety of conditions encountered by magma during ascent, we propose expanding the  
14 model, developed in this study for isothermal decompression, to a wider range of pressures and  
15 temperatures (i.e., non-isothermal conditions). We present new measurements of feldspar growth rates  
16 during decompression in water-saturated rhyolite, emphasizing the importance of accurately calculating  
17 the nucleation delay in order to estimate growth rates. Using these data, we propose a method for  
18 estimating the ascent rate of water-saturated rhyolite based on the nucleation delay of feldspar microlites.  
19 The findings of this study hold the promise of a better understanding of volcanic timescales during  
20 magma ascent in the conduit, as well as the possibility of extending our model to more complicated  
21 scenarios with changing pressure and temperature.

22 **Keywords:**

23 nucleation delay, ascent rate, crystallization, decompression, magmatic timescales

24 **Introduction**

25 Microlites, the crystals less than few hundred micrometers in size, of which plagioclase is the most  
26 common, form on the timescales of minutes to a few days during magma ascent and subsequent degassing  
27 (e.g., Sharp et al. 1996, Hammer et al. 1999, Hammer and Rutherford 2002, Martel 2012, Mollard et al.  
28 2012). Because they form on the timescales of ascent their presence and textures can provide valuable  
29 information about magma ascent rates and conditions in the conduit. Moreover, timing of crystallization  
30 can affect the production of crystal-rich or crystal-poor magmas, resulting in changes in the textural  
31 make-up of the produced rock as well as the explosivity. Magma composition and exsolution of dissolved  
32 volatiles are the primary determinants of eruption style and timing. The presence of crystals, on the other  
33 hand, can impact volatile exsolution by acting as nucleation sites for bubble nucleation (e.g., Hurwitz  
34 and Navon 1994, Gardner and Denis 2004, Shea 2017, Pleše et al. 2018) and magma rheology (D'Oriano  
35 et al. 2005, Mader et al. 2013, Vona et al. 2017). The permeability of vesicular magma can be affected  
36 by crystallization during ascent, which can lead to changes in eruption style by lowering the percolation  
37 threshold and changing the eruptive behaviour from explosive to effusive (e.g., Lindoo et al. 2016,  
38 Cassidy et al. 2018, deGraffenried et al. 2019). Notably, magmas with ~4.0-5.5 wt. % water (around the  
39 water saturation limit depending on storage conditions) and <30 vol. % crystallinity erupt mostly  
40 explosively, but magmas with >40 vol. % crystallinity erupt primarily effusively (Popa et al. 2021).

41 The first order transition i.e., nucleation initiates the process of partial or complete crystallization of a  
42 silicate melt (e.g., Kirkpatrick 1981, De Yoreo and Vekilov 2004, Fokin et al. 2006). Nucleation is the  
43 process of rearrangement of atoms (or molecules) in a parent phase that results in the formation of a  
44 cluster of a new, thermodynamically more stable phase (Kirkpatrick 1981, De Yoreo and Vekilov 2004,  
45 Fokin et al. 2006). In magmatic systems, however, nucleation does not occur immediately when the new  
46 phase becomes thermodynamically stable, instead can be delayed by minutes, hours, or days (e.g., Fenn  
47 1977, Swanson 1977, Couch 2003, Mollard et al. 2012, Polacci et al. 2018, Rusiecka et al. 2020).

48 Understanding the magmatic timescales of volcanic systems can be improved by studying nucleation  
49 kinetics, particularly nucleation delay. Modelling nucleation delay allows for the prediction of the

50 timescales on which igneous rocks crystallize from a melt and when they do remain vitreous (Rusiecka  
51 et al. 2020). Numerous studies have been conducted to investigate crystallization kinetics in felsic melts  
52 (e.g., Fenn 1977, Swanson 1977, Hammer and Rutherford 2002, Couch 2003, Brugger and Hammer  
53 2010, Mollard et al. 2012, Riker et al. 2015, Sirbescu et al. 2017, Mollard et al. 2020, Rusiecka et al.  
54 2020). However, the majority of these studies have been focused on constraining the crystal growth and  
55 nucleation rates. A few of them provided incidental measurements of nucleation delays (e.g., Fenn 1977,  
56 Swanson 1977, Couch 2003, Mollard et al. 2012, Sirbescu et al. 2017). Rusiecka et al. (2020) presented  
57 a model based on Classical Nucleation Theory (CNT) for the nucleation delay of plagioclase, pyroxene  
58 and olivine in basaltic melts, and of quartz and alkali feldspar in a hydrous, rhyolitic melt under isobaric  
59 conditions.

60 In this contribution we build on the model presented by Rusiecka et al. (2020) by developing a theoretical  
61 model of feldspar nucleation delays in rhyolitic melts during ascent in the conduit. The model was  
62 calibrated by a series of decompression-induced crystallization experiments conducted using a hydrated  
63 obsidian at temperatures ranging from 800-900 °C and pressures ranging from 25-200 MPa. This  
64 temperature range is representative of natural rhyolite temperatures (e.g., Castro and Dingwell 2009) and  
65 decompression from 200 to 25 MPa covers the range of pressures prevailing during magma ascent from  
66 a deep reservoir (200 MPa, around 7.5 km below the Earth's surface) to near-surface levels (Martel, 2012).

### 67 **Classical Nucleation Theory**

68 Volmer and Weber (1926), Becker and Döring (1935), and Turnbull and Fisher (1949) developed classical  
69 nucleation theory during the first half of the twentieth century on the basis of Gibbs' (1875-1878) earlier  
70 work on the thermodynamic description of heterogeneous systems. According to Gibbs' theory, a  
71 heterogeneous system, consisting of a parent phase and a new nucleating phase, is replaced for simplicity,  
72 by two homogeneous phases (the parent and a nucleus) divided by a mathematical surface of zero  
73 thickness. A change in Gibbs free energy between the newly formed nucleus and the parent phase drives  
74 nucleation. The parent phase is assumed to be in a metastable state, and nucleation must occur after an

75 activation energy barrier is overcome. The rate of nucleation is affected by the degree of supersaturation,  
76 which is commonly measured as undercooling below the liquidus temperature. When the new nuclei are  
77 surrounded by the previously existing phase, homogeneous nucleation occurs, and heterogeneous  
78 nucleation occurs when the nucleating phase forms on a previously existing surface (Kirkpatrick 1981,  
79 De Yoreo and Vekilov 2004, Fokin et al. 2006).

## 80 **Methods**

### 81 **Experimental procedure**

82 The starting sample used in the experiments is the Lake County Obsidian (LCO) – a common  
83 metaluminous composition (in wt. % on an anhydrous basis: 75.7 SiO<sub>2</sub>, 0.1 TiO<sub>2</sub>, 13.2 Al<sub>2</sub>O<sub>3</sub>, 0.7 FeO,  
84 0.1 MnO, 0.1 MgO, 0.6 CaO, 4.3 Na<sub>2</sub>O, 5.1 K<sub>2</sub>O; Maneta and Baker 2014). This is the same material as  
85 used to investigate cooling-induced crystallization by Rusiecka et al. (2020) and Rusiecka and Baker  
86 (2021). The obsidian was coarsely crushed and melted at 1400 °C for 4 hours before being ground to a  
87 fine powder. About 30 mg of the LCO glass powder was loaded into 15 mm length 2.9 mm OD (outer  
88 diameter), 2.5 mm ID (inner diameter) gold capsules with enough water to ensure saturation (as  
89 calculated using MagmaSat, Ghiorso and Gualda 2015) and welded shut. To check for leaks, the capsules  
90 were kept at 120 °C for at least 2 hours and weighed. The experiments were carried out at the Institut des  
91 Sciences de la Terre d'Orleans in Orleans (France) in an Internally Heated Pressure Vessel (IHPV) with  
92 argon as the pressurizing medium. The capsules were placed in the 3-cm hot spot of the furnace and  
93 temperature ( $\pm 4$  °C) was controlled at the top and bottom of the samples using two S-type thermocouples.  
94 The pressure ( $\pm 2.5$  MPa) was monitored by a high pressure transducer calibrated against a 700 MPa  
95 Heise gauge. The liquidus temperature at different pressures was determined using rhyolite-MELTS  
96 (Ghiorso and Sack 1995, Gualda et al. 2012) and confirmed experimentally at 200, 150 and 50 MPa to  
97 constrain the effective degree of undercooling ( $\Delta T_{\text{eff}} = \text{run temperature} - \text{liquidus temperature}$ , Table 1).  
98 To confirm that the starting material was free of pre-existing crystals we conducted an experiment in  
99 which the samples were kept for 24 h at 825 °C and 200 MPa and then decompressed at 10 MPa/min to

100 50 MPa. They were then immediately isobarically quenched. Experiments were first kept at 200 MPa  
101 and 825 °C for 24 hours to homogenize the melt with water and then decompressed at 10 MPa/min to  
102 90, 75, 50 and 25 MPa (Table 1).

### 103 **Analytical techniques**

104 After the experiments, the capsules were first weighted to confirm that no water loss occurred during the  
105 experiment. Then they were opened, and the samples were recovered as glass chips that were placed in  
106 epoxy resin and polished for analyses. The samples were imaged using a Merlin Compact Zeiss scanning  
107 electron microscope (SEM) at the Institut des Sciences de la Terre d'Orléans in Orléans. Although most  
108 of the crystals were too small for thorough quantitative analysis the observed crystals were identified as  
109 plagioclase feldspar (i.e., in the oligoclase field in the experiment conducted at 50 MPa; semi-quantitative  
110 composition:  $An_{28}Ab_{65}Or_7$ ) and magnetite using a Bruker Quantax XFlash6 energy dispersive  
111 spectrometer coupled to the SEM. The crystallinity of the samples did not exceed ~1-2 vol. %. The water  
112 saturation content was calculated using the MagmaSat application, which employs the Ghiorso and  
113 Gualda (2015)  $H_2O-CO_2$  saturation model.

### 114 **Theoretical model**

115 Nucleation delay ( $\tau$ ) is the time between when the parent phase (silicate melt) is cooled below its liquidus  
116 temperature and the onset of crystallization. It is defined as:

$$117 \tau = \frac{16h}{\pi} \frac{\sigma}{\Delta G_V^2 a^4} \exp \frac{E_a}{RT} \quad (1)$$

118 where  $h$  is Planck's constant,  $R$  is the gas constant,  $\sigma$  is the interfacial free energy between the parent  
119 phase and the forming nucleus. The value for the interfacial free energy was chosen to be 0.18 J/m<sup>2</sup>, this  
120 being the value of the interfacial free energy for silicate crystals in the system  $Na_2O-CaO-SiO_2$  (Fokin et  
121 al. 2006). The interfacial free energy, this being  $\sigma$ , is poorly constrained and for feldspar in a silicate melt  
122 can vary between 0.10 and 0.25 J/m<sup>2</sup> (e.g., Hammer 2004, Baker et al. 2020, Mollard et al. 2020, Rusiecka  
123 et al. 2020). Free energy of crystallization per unit volume  $\Delta G_V$  (Robie and Bethke 1962) was calculated

124 using the MELTS software (Ghiorso and Sack 1995, Gualda et al. 2012). Energy of crystallization,  $\Delta G$   
125 was obtained using the affinities of feldspar in the melt of the same composition as the starting material  
126 at the temperature of the experiments (825 °C) and a range of pressures (10 to 200 MPa). The affinities  
127 were plotted against pressure and fitted with a trend line. The equation describing the trend line was used  
128 in the calculation of the  $\Delta G$  in the nucleation delay program. Parameter  $a$  in Equation (1) is the size of a  
129 structural unit, here approximated using the size of the  $\text{Si}^{4+}$  ion ( $0.26 \times 10^{-10}$  m, Shannon 1976), as  $\text{Si}^{4+}$   
130 is an essential component of both a silicate melt and its crystals. This value was previously successfully  
131 used by Rusiecka et al. (2020) for nucleation of quartz and feldspar in a natural, hydrous rhyolite, and by  
132 Baker et al. (2020) for anorthite and diopside in a synthetic composition. The activation energy for  
133 nucleation ( $E_a$ , the kinetic barrier) was approximated using the diffusivity calculated using the Eyring  
134 equation (Eyring 1935):

$$135 \quad E_a = -k_B T \ln \left( \frac{h}{\lambda^3 \eta} \right) \quad (2)$$

136 where  $\eta$  is viscosity and  $\lambda$  is the size of the diffusive jump and  $k_B$  is the Boltzmann's constant. The  
137 diffusive jump is not well constrained and can generally vary between  $1^{-10}$  to  $5^{-10}$  m (0.1 to 0.5 nm, e.g.,  
138 Baker 1990, 1992, Mungall 2002, Gonzalez-Garcia et al. 2017). The variation of the diffusive jump from  
139  $1^{-10}$  m to  $5^{-10}$  m corresponds to variation in a nucleation delay of two orders of magnitude. The viscosity  
140 for the Eyring equation was calculated using model by Giordano (2008) with water contents calculated  
141 at saturation at the different run pressures using the MagmaSat program (i.e., 1.8 to 4.8 wt. %  $\text{H}_2\text{O}$  from  
142 25 to 120 MPa). Rusiecka et al. (2020) proposed that activation energy for the nucleation needs to be  
143 modified by using diffuse interface theory. However, this is not the case in this study as using the  
144 parameters for water saturated melt and changing the water content at saturation with pressure replicates  
145 the experimental results to within a factor of two. This variation might be caused by the differences in  
146 water content and the distribution of water in the melt. In the case of Rusiecka et al. (2020) water content

147 was 4 wt. %, which is several wt. % below the saturation content at the experimental conditions (600  
148 MPa) whereas in this study all the experiments were water saturated.

149 The numerical constant in Equation 1 ( $\frac{16}{\pi}$ ) is that of Fokin et al. (2006); different derivations of Equation  
150 (1) (e.g., Collins 1955, Kashchiev 1969, Slezov and Schmelzer 1999, 2002) can lead to slightly different  
151 constant values, but they typically vary by less than an order of magnitude (Gutzow and Schmelzer 1995).

## 152 **Results**

### 153 **Description of the textural characteristics of the experimentally produced plagioclase**

154 All samples produced in experiments conducted in this study are vesicular and contain magnetite.  
155 Although magnetite may be seen as inclusion in some feldspars on the SEM images, it is not considered  
156 here as a nucleation site for the silicate crystals for two main reasons. Firstly, the distribution of the oxides  
157 in connection to the plagioclase is random (i.e., there are more oxide sites than plagioclase crystals).  
158 Secondly, the magnetite distribution within the plagioclase crystals is random (i.e., magnetite is not  
159 systematically in the centre of the plagioclase where growth would presumably have started in case of  
160 magnetite acting as a preferential nucleation site). The sub-micron oxides were likely included in the  
161 plagioclase during rapid growth. Plagioclase was produced at three different pressures: 90, 75 and 50  
162 MPa (corresponding to  $\Delta T_{\text{eff}} = -10, -20$  and  $-55$  °C). The representative images of the samples produced  
163 at those conditions can be observed in Figure 1. Magnetite and plagioclase crystals are homogeneously  
164 distributed in the sample and did not preferentially nucleated at capsule walls. At 50 MPa ( $\Delta T_{\text{eff}} = -55$  °C)  
165 the crystals are highly irregular with skeletal morphologies (Fig. 1a) and numerous embayments and melt  
166 inclusions (Fig. 1b). At 75 MPa ( $\Delta T_{\text{eff}} = -20$  °C) the crystals are more regular than at 50 MPa and have  
167 more prismatic shapes, nevertheless some irregularities, melt embayments and inclusions persist (Fig.  
168 1c). At 90 MPa ( $\Delta T_{\text{eff}} = -10$  °C) the crystals are elongated, with euhedral and subhedral morphologies,  
169 well faceted crystals (Fig. 1d), and some clustering of smaller crystals.

### 170 **Nucleation delay**



171 The theoretically derived curve describing nucleation delay of plagioclase in hydrous, metaluminous  
172 rhyolite at the conditions of the experiments in this study is shown in Figure 2 along with the experimental  
173 results. In our study, the shortest delay is 30 h at  $\Delta T_{\text{eff}} -50$  °C, and the delay tends to infinity as the  
174 undercooling approaches zero. The findings of our study, as well as our modeling of the nucleation delay  
175 in decompression induced crystallization, were compared to those of Mollard et al. (2012), who  
176 conducted experiments on a rhyolitic melt using a crystal free starting material and reported nucleation  
177 delays in their study (nucleation delays are given in Fig. 7 of Mollard et al. 2012). We initialized  
178 nucleation delay model with a value for interfacial free energy of  $0.18 \text{ J/m}^2$ , and bracketed it with values  
179 ranging from  $0.10 \text{ J/m}^2$  to  $0.25 \text{ J/m}^2$ . Our theoretical modelling is specific to the composition used in this  
180 study, but agrees well with other rhyolitic compositions represented by the synthetic composition of  
181 Mollard et al. (2012), as shown here in Figure 2. Considering both the experiments conducted in this  
182 study as well as the experiments of Mollard et al. (2012), the model agrees with the experimental data  
183 by a factor of two, which is also the uncertainty represented by the typical variation in range of the  
184 interfacial free energy.

### 185 **Growth rates**

186 We calculated the growth rates of feldspar ( $G_{\text{max}}$ ) in the samples where crystallization was observed. To  
187 do this we used ten largest crystals from each sample and applied growth rate equation (e.g., Couch 2003,  
188 Arzilli and Carroll 2013, Arzilli et al. 2020, Rusiecka and Baker 2021) modified to include nucleation  
189 delay:

$$190 \quad G_{\text{max}} = \frac{(L/2)}{(t-\tau)} \quad (3)$$

191 Here, L is the length of the longest crystal dimension (in m), t is the duration of the experiment (in s) and  
192  $\tau$  is the nucleation delay calculated using the model described above. The spreadsheet with all the growth  
193 rates obtained is available in the Supplementary Material.

194 Growth rates for pressures of 90, 75 and 50 MPa (corresponding to  $\Delta T_{\text{eff}} = -10, -20$  and  $-55$  °C) are shown  
195 in Figure 3. At the conditions of these experiments (i.e., moderate to low degrees of undercooling) the  
196 growth rates are on the order of  $10^{-11}$  m/s ( $10^{-8}$  mm/s), and increases from the lowest degree of  
197 undercooling to the highest degree of undercooling.

198 When plotting the growth rates for experiments conducted at 50 MPa ( $\Delta T_{\text{eff}} = -55$  °C) with different  
199 durations (48, 72 and 120 h, Fig. 4), higher by about a factor of two in the first, shorter experiment than in  
200 the subsequent, longer experiment. Growth rates decrease with time is not unexpected. We expect non-  
201 linear behaviour due to the rapid decay of initial steep gradients in chemical potential (c.f., Moschini et  
202 al. 2021). The decay in the growth rates can be best represented semi-quantitatively by fitting the  
203 experimental results with a trend line, as in Figure 4.

204 Another way of looking at growth rates is by comparing them to the equilibrium growth rate ( $G$ ) as  
205 calculated using the dependence of growth rate on the degree of undercooling (Equation 4; Toramaru  
206 1991, Moschini et al. 2021):

$$207 \quad G \propto \exp\left(\frac{-E_a}{RT_{\text{exp}}}\right) \left(1 - \exp\left(\frac{\Delta H \Delta T}{RT_{\text{exp}} T_L}\right)\right) \quad (4)$$

208 Here,  $E_a$  is the activation energy (approximated as the activation energy of diffusion from Eq. 2 with the  
209 same parameters as in the nucleation delay calculation),  $T_{\text{exp}}$  is the temperature at which the experiment  
210 was conducted,  $\Delta H$  is enthalpy of fusion (extracted from MELTS), and  $T_L$  is the liquidus temperature.  
211 Growth rates calculated using Equation (4) yield results on the same order of magnitude as the growth  
212 rates derived from experimental results with nucleation delay taken into account. Moreover, the growth  
213 rate at 50 MPa most closely matches the growth rate derived from the longest experiment suggesting that  
214 after dissipation of the initial steep chemical potential gradients, the growth rate trends to equilibrium  
215 over time. In the case of growth of plagioclase, at 50 MPa the equilibrium growth rate is achieved after  
216 ~95 h (120 h of the experimental duration minus ~25 h for the nucleation delay).

217 **Discussion**

218 **Using nucleation delay to estimate the ascent rate in microlite-free and microlite-bearing rhyolites**

219 Textures of volcanic products are strongly linked to the history of magma ascent (e.g., Noguchi et al.  
220 2006, Polacci et al. 2006, Bain et al. 2021) . On eruptive timescales, degassing of volatile-rich magma  
221 can induce feldspar microlite formation, although this growth can be hindered if the ascent is too fast  
222 (c.f., Hammer and Rutherford 2002). For volcanic hazard reduction, estimating the ascent rates during  
223 an eruptions is crucial, as it can determine the style of eruption (e.g., Dingwell, D.B. 1996, Castro and  
224 Gardner 2008, Mujin and Nakamura 2014). Typical ascent rates derived for rhyolitic eruptions are in the  
225 range of 0.00005 to 5 MPa/s, corresponding to ascent velocities of 0.0001 to 200 m/s (Rutherford 2008).  
226 Figure 5 presents the comparison between depressurization equivalent ascent rates of 0.0005 MPa/s,  
227 0.001 MPa/s, 0.005 MPa/s and 0.01 MPa/s, and the nucleation delay of feldspar in water saturated  
228 rhyolite. The starting pressure for the ascent paths, simulating the initial storage conditions, is 120 MPa.  
229 For the composition considered at 825 °C these conditions are above the liquidus. The heterogeneous  
230 nature of nucleation requires using the correction factor ( $\tau_{het}$ ):

231 
$$\tau_{het} = \tau\varphi^{\frac{1}{3}}$$
 (5)

232 where

233 
$$\varphi = \frac{1}{2} - \frac{3}{4}\cos\theta + \frac{1}{4}\cos^3\theta$$
 (6)

234 here  $\theta$  is the interior contact angle between the surface upon which nucleation occurs and the  
235 nucleating phase (Gutzow and Schmelzer 1995; Fokin et al. 2006). The angle used here (25°) is the  
236 median wetting angle for feldspar crystals in rhyolite (Holness 2006). In the case of homogeneous  
237 nucleation, none of the ascent paths cross the nucleation delay curve except for the path with an ascent  
238 rate of 0.0005 MPa/s. The presence of microlites in a water-saturated rhyolite under conditions  
239 considered in this study would mean that if the ascent started at the superliquidus storage conditions of  
240 120 MPa and 825 °C, then the minimum ascent rate would be between 0.0005 and 0.001 MPa/s. If no  
241 feldspar microlites were produced, the maximum ascent rate would be 0.001 MPa/s. Presence of pre-

242 existing crystals in the melt subjected to decompression may influence the nucleation kinetics during  
243 ascent by decreasing the interfacial free energy between the parent phase (melt) and new nuclei,  
244 consequently decreasing the nucleation delays (c.f., Martel 2012, Rusiecka et al. 2020). In case of  
245 heterogeneous nucleation delay (Fig. 5) the ascent path crosses the nucleation delay curve at 0.005  
246 MPa/s and not at 0.01 MPa/s, suggesting that in the absence of microlites or overgrowths on pre-  
247 existing crystals the maximum ascent rate was 0.01 MPa/s. If microlites or overgrowths are present the  
248 minimum ascent rate declines, for this case, to 0.005 MPa/s. This simple model makes it possible to  
249 estimate the maximum ascent rate of the crystal free rhyolite, and the minimum ascent rate of  
250 microlite-bearing rhyolite.

251 An example of how presence or absence of microlites can be used to estimate the ascent rate is the  
252 2008 eruption of Chaitén (Northern Patagonia, Chile). There has been discussion as to the reason of the  
253 lack of microlites in the products of this eruption (Andrews and Befus 2020, Huggins et al. 2021).  
254 Andrews and Befus (2020), who in presenting their Supersaturation Nucleation and Growth of  
255 Plagioclase (SNGPlag) numerical model, suggested that the reason their model was unable to  
256 reproduce the crystal poor textures of the 2008 Chaiten eruption was the fact that they did not include  
257 nucleation delay in their calculations. Castro and Dingwell (2009) performed decompression  
258 experiments to determine the minimum decompression rate which resulted in the textures observed in  
259 the products of the 2008 eruption. They observed that decompression at 10 MPa/h and 20 MPa/h  
260 ( $\sim 0.005$  MPa/s) produced an overgrowth on pre-existing plagioclase crystals but the rim did not form  
261 as a result of decompression at 40 MPa/h ( $\sim 0.01$  MPa/s). This result agrees well with the model  
262 presented here, where the nucleation delay curve for heterogeneous nucleation crosses the ascent path  
263 at 0.005 MPa/s suggesting formation of plagioclase at this ascent rate, but not at 0.01 MPa/s. This  
264 indicates that the ascent of 0.01 MPa/s is too fast for the plagioclase to form.

265 **Modelling nucleation delay in Pressure-Temperature (PT) space**

266 Because just one isothermal decompression scenario is used, the nucleation delay model described above  
267 may have limited utility in calculating nucleation delays of feldspar in rhyolitic compositions across a  
268 wide range of conditions. However, the successful comparison of the nucleation delay model to the  
269 decompression results of this study, and the isobaric tests of Rusiecka et al. (2020), allow for the  
270 construction of a model of nucleation delay change with pressure and temperature. The nucleation delay  
271 at different pressures and temperatures was calculated using a 40 by 40 grid of P and T with a pressure  
272 step of 25 bars (2.5 MPa). This corresponds to the uncertainty of the pressure in the experiments and the  
273 temperature step of 5 °C. Figure 6 shows the results of modelling of nucleation delay from 100 MPa to  
274 10 MPa and for temperatures from 800 °C to 650 °C. The highest nucleation delays are observed close  
275 to the liquidus, at high pressure and temperature, and at the highest degree of undercooling, at low  
276 pressure and temperature. At low pressure and temperature there is a high degree of undercooling and  
277 the nucleation delay is primarily controlled by the activation energy. Therefore the barrier preventing  
278 nucleation is mainly kinetic. At high pressure and temperature, close to the liquidus, the nucleation is  
279 mostly hindered by the thermodynamic barrier, i.e., the interplay between the surface and bulk free  
280 energy.

281 Our findings may expand the use of nucleation delay and crystallization of microlites into more  
282 complicated magma ascent scenarios, for example: one in which the temperature is not constant during  
283 decompression. Thermodynamic modelling of magma degassing without crystallization indicates that  
284 both heating and cooling is possible (c.f., Mastin and Ghiorso 2001, Glazner 2019). Not only can the  
285 degassing result in a change in temperature during decompression, but also the latent heat released during  
286 crystallization of phases not considered in the nucleation delay model may increase the temperature of  
287 the magma (c.f., Couch et al. 2001, Blundy et al. 2006). Such phases include those that may nucleate  
288 before plagioclase, such as Fe-Ti oxides, pyroxene, and amphibole. The results from our modelling are  
289 available in Supplementary material.

290 **Re-evaluating feldspar growth rates**

291 Growth rates when the nucleation delay is considered are twice to three times faster than when the growth  
292 is considered throughout the whole experiment (Fig. 3). The same can be observed when comparing  
293 growth rates obtained at 50 MPa and for different durations, with the largest difference being visible  
294 when comparing growth rates for the shortest experiments (Fig. 4). Growth rates calculated with and  
295 without consideration of the nucleation delay have been fitted with an exponential trend as described in  
296 Figure 4. The trend line for growth rates calculated without nucleation delay has the same, but the  
297 intercept two times lower.

298 It is especially important to account for the nucleation delay when the timescales being considered are  
299 on the same order of magnitude as the delay. This is the case for our experiments, as well as when  
300 considering the formation of microlites in an ascending magma where timescales are short i.e., velocities  
301 and decompression rates are fast. Experimental growth rates adjusted for delayed nucleation and applied  
302 to volcanic systems, can thus help to constrain the minimum ascent rate based on the presence and size  
303 of the microlites. When calculating growth rates we need to, however, not only consider the delayed  
304 nucleation, but also the deviation from the equilibrium growth rate.

305 Growth rates obtained in this study fall within the range of growth rates determined in previous studies  
306 on decompression induced crystallization in felsic melts (e.g., Hammer and Rutherford 2002, Couch  
307 2003, Martel 2012, Mollard et al. 2012, Riker et al. 2015). Our growth rates are compared to the results  
308 of other experimental studies on silicic melts in which the single step decompression (SSD) method was  
309 employed in (Fig. 3).

310 Mollard et al. (2012) obtained the growth rates one order of magnitude higher than in our study at similar  
311  $\Delta T_{\text{eff}}$  whereas the growth rates in Hammer and Rutherford (2002) and Couch (2003) are of the same order  
312 of magnitude as obtained here. One reason for this discrepancy may be the effect of composition. This  
313 study and the studies of Hammer and Rutherford (2002) and Couch (2003) used compositions with  $\text{SiO}_2$   
314 contents of less than 75 wt. % and containing  $\text{K}_2\text{O}$ . Mollard et al. (2012) used a high silica composition  
315 ( $\text{SiO}_2 = 78.7$  wt. %) and with no  $\text{K}_2\text{O}$ . The growth rates of Hammer and Rutherford (2002) and Couch

316 (2003) are, however, consistently lower than those obtained here (with exception of two short duration  
317 experiments of Couch 2003). However, the results of Hammer and Rutherford (2002) and Couch (2003)  
318 fall within the uncertainty of our growth rates calculated without taking the nucleation delay into  
319 consideration. This further strengthens the importance of accurately estimating nucleation delays when  
320 trying to obtain and compare growth rates.

## 321 **Conclusions**

322 Theoretical (Classical Nucleation Theory) and experimental nucleation delays for decompression-  
323 induced crystallization of feldspar in rhyolitic melt agree within a factor of two. This finding opens up  
324 the possibility of quantitatively predicting feldspar nucleation during decompression in water-saturated  
325 rhyolite. This, thus, provides an opportunity to better understand the timescales of ascent, decompression  
326 and nucleation. As part of this, nucleation delay is particularly important to consider for accurate growth  
327 rate estimation, particularly under disequilibrium conditions resulting in rapid growth. Nucleation delay  
328 can be used to calculate the maximum ascent rate of microlite-free rhyolites and the minimum ascent rate  
329 of microlite-bearing rhyolites. We thus extend the simple model for cooling-induced and decompression-  
330 induced crystallization, allowing the nucleation delay of feldspar in rhyolites to be estimated under a range  
331 of eruptive conditions in shallow volcanic systems.

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540 **Figure and Table Captions:**

541 **Figure 1.** BSE (back-scattered electron) images of the experimental run products at a, b) 50 MPa ( $\Delta T_{\text{eff}}$   
542 = -55 °C): glass with skeletal feldspar crystals and magnetite, in b) several melt inclusions can be  
543 observed; c) 75 MPa ( $\Delta T_{\text{eff}}$  = -20 °C) – glass with feldspar and magnetite crystals and visible vesicle; d)  
544 90 MPa ( $\Delta T_{\text{eff}}$  = -10 °C) well faceted feldspar crystals in glass with magnetite e) low magnification BSE  
545 image showing one of the glass chips recovered after the experiment

546 **Figure 2.** Nucleation delay of plagioclase in metaluminous rhyolite, empty symbols – no crystals  
547 observed, filled symbols – crystals observed, circles – this study, squares – Mollard et al. 2012, solid  
548 black curve – nucleation delay calculated using Eq. 1. dotted and dashed lines – nucleation delay curves  
549 calculated using the range of interfacial free energy of 0.10 to 0.25 J/m<sup>2</sup>. Pressure relates to this study,  
550 while  $\Delta T_{\text{eff}}$  relates to both this study and the study of Mollard et al. 2012.

551 **Figure 3.** Growth rates calculated using Eq. 3 at various experimental conditions for average of 10 largest  
552 crystals for each samples, circles – this study, squares – Couch 2003, stars – Hammer and Rutherford  
553 2002; Pressure relates to this study, while  $\Delta T_{\text{eff}}$  is relevant for all the presented studies.

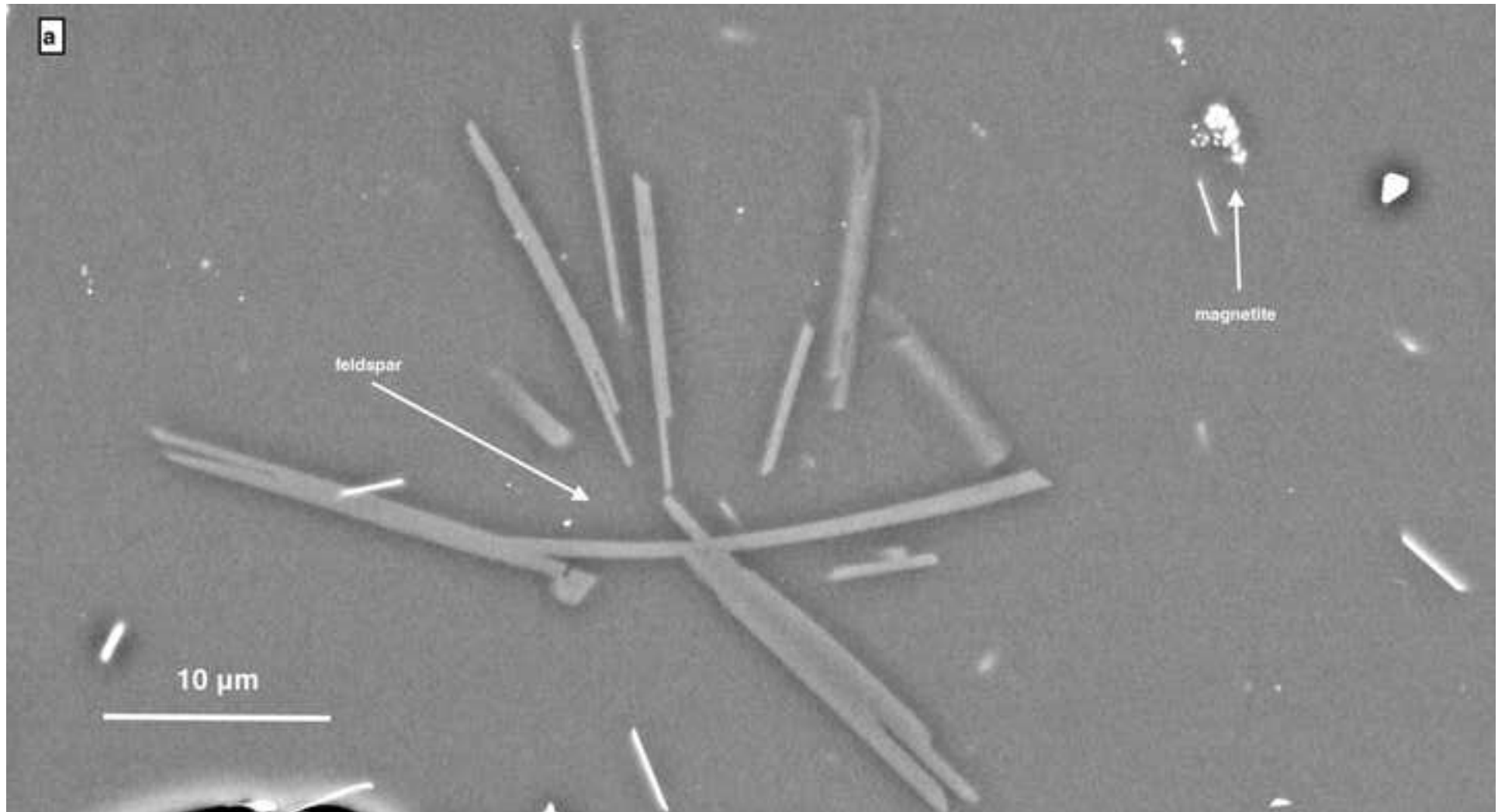


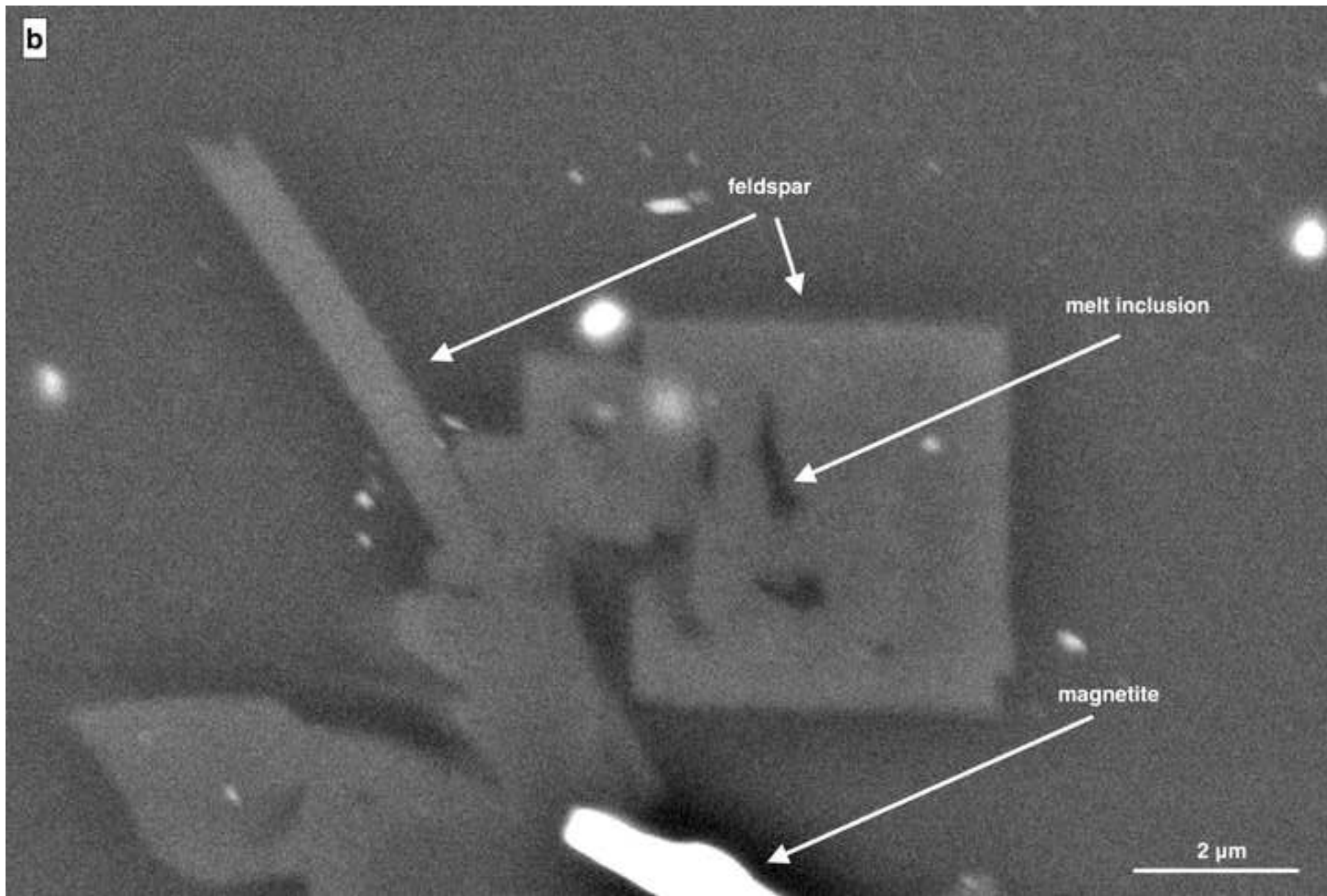
554 **Figure 4.** Growth rates calculated using Eq. 3 at 50 MPa at various experimental durations (48, 72 and  
555 120 h) for average of 10 largest crystals for each sample, black symbols – growth rates calculated with  
556 taking nucleation delay into consideration, grey symbols – growth rates calculated without taking  
557 nucleation delay into consideration, dashed line – exponential trend line fitted to the data.

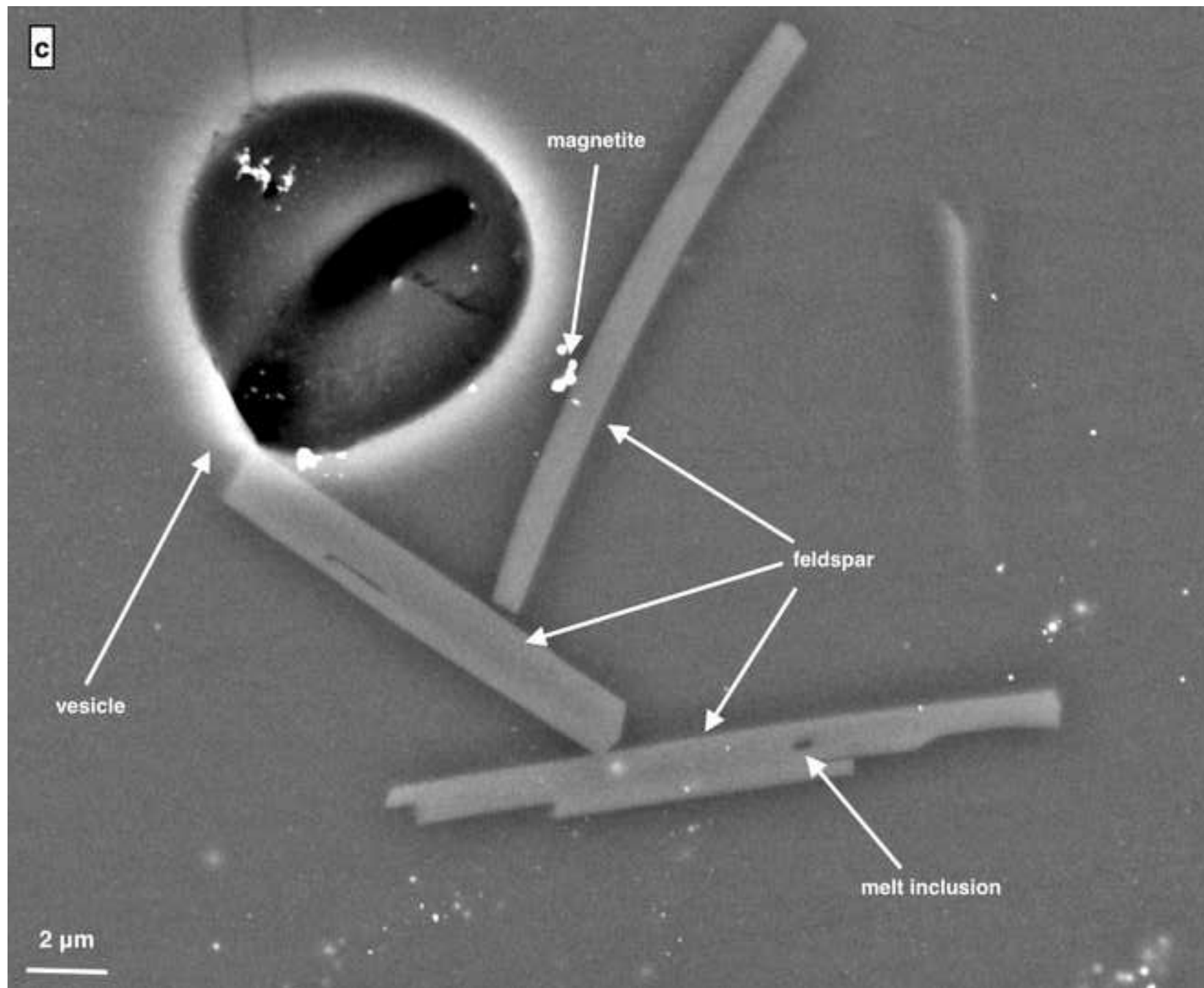
558 **Figure 5.** Nucleation delay curves for homogeneous (solid black curve) and heterogeneous (dotted black  
559 curved) nucleation of feldspar calculated using the theoretical model for feldspar nucleation in water-  
560 saturated rhyolite, grey dashed lines – ascent paths.

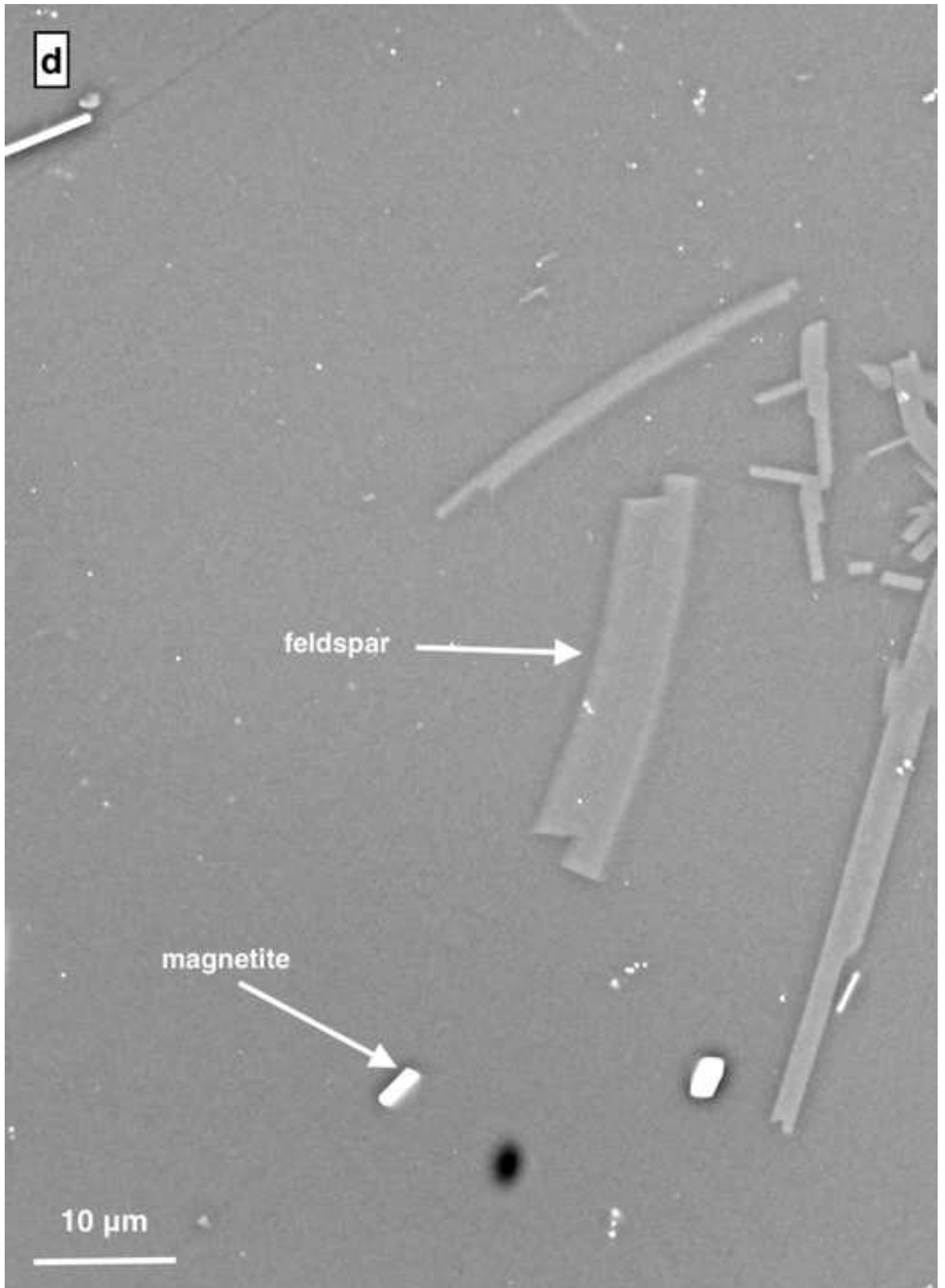
561 **Figure 6.** Nucleation delay calculated in the PT space for water-saturated rhyolite with the model and  
562 parameters described in the text of the manuscript. Colormap ‘hawaii’ (Crameri 2018, Crameri et al.  
563 2020) was used to ensure accessibility for readers.

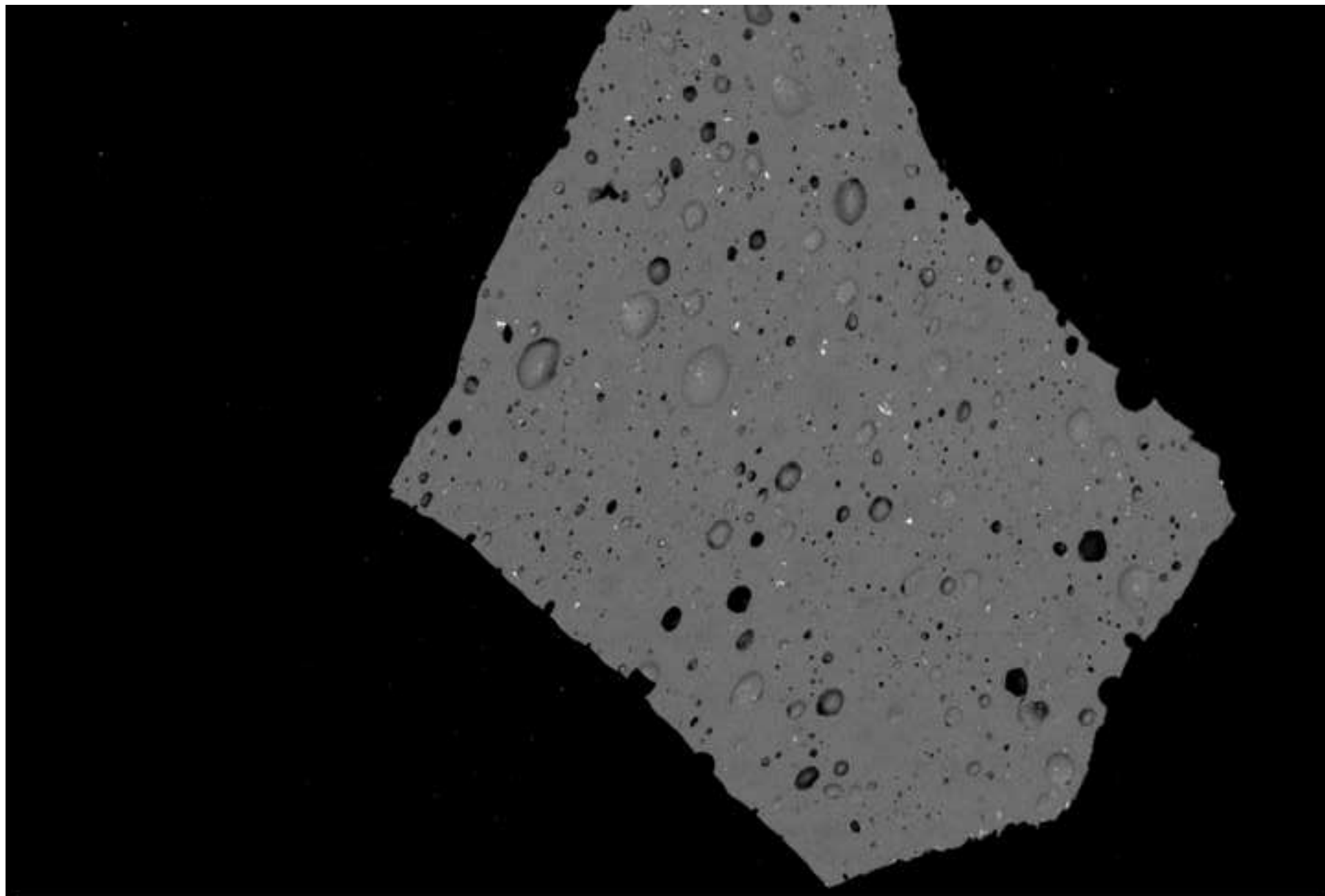
564 **Table 1.** Experimental run table, LCOL – experiments conducted to confirm the location of the liquidus  
565 calculated using MELTS, LCOD – decompression single step decompression (SSD) experiments;  $P_i$  –  
566 initial pressure,  $P_f$  – final pressure,  $\Delta T_{\text{eff}}$  – effective undercooling,  $dP/t$  – rate of decompression, gl –  
567 glass, mgt – magnetite, fds – feldspar.











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200 μm  
|-----|

EHT = 15.00 kV  
Mag = 33 X

Signal A = BSD4 A  
WD = 11.0 mm

Width = 3.465 mm



Figure 2

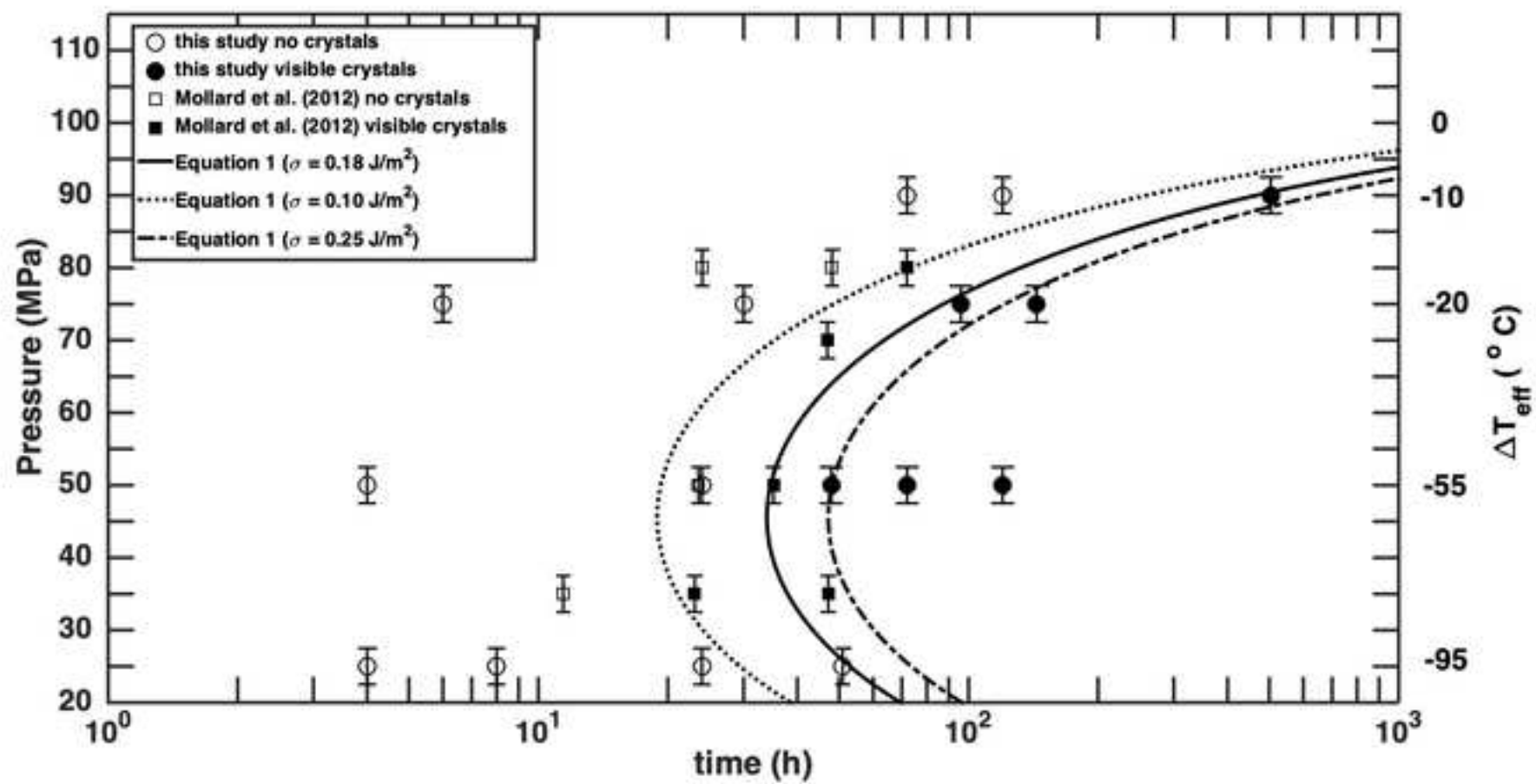
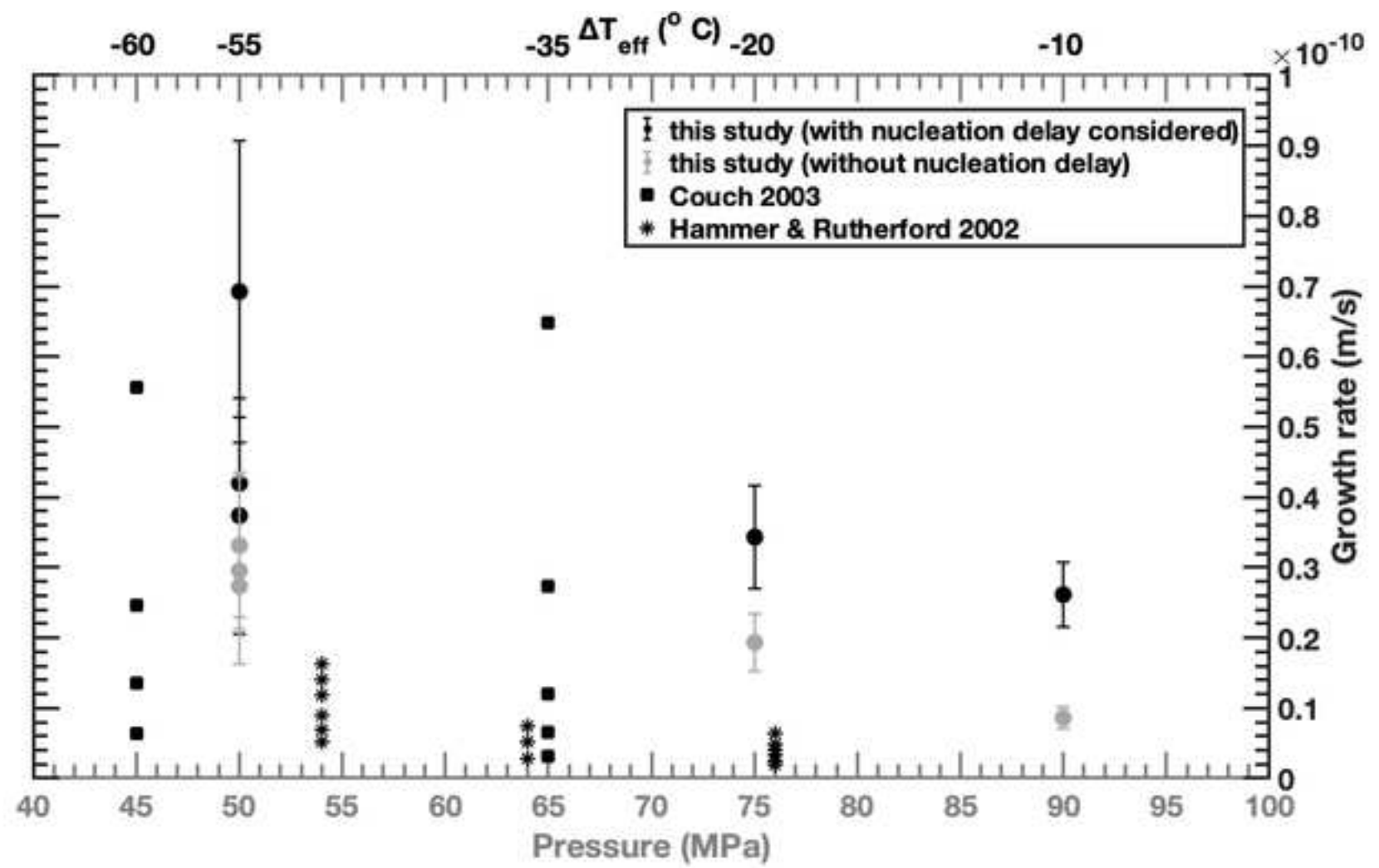
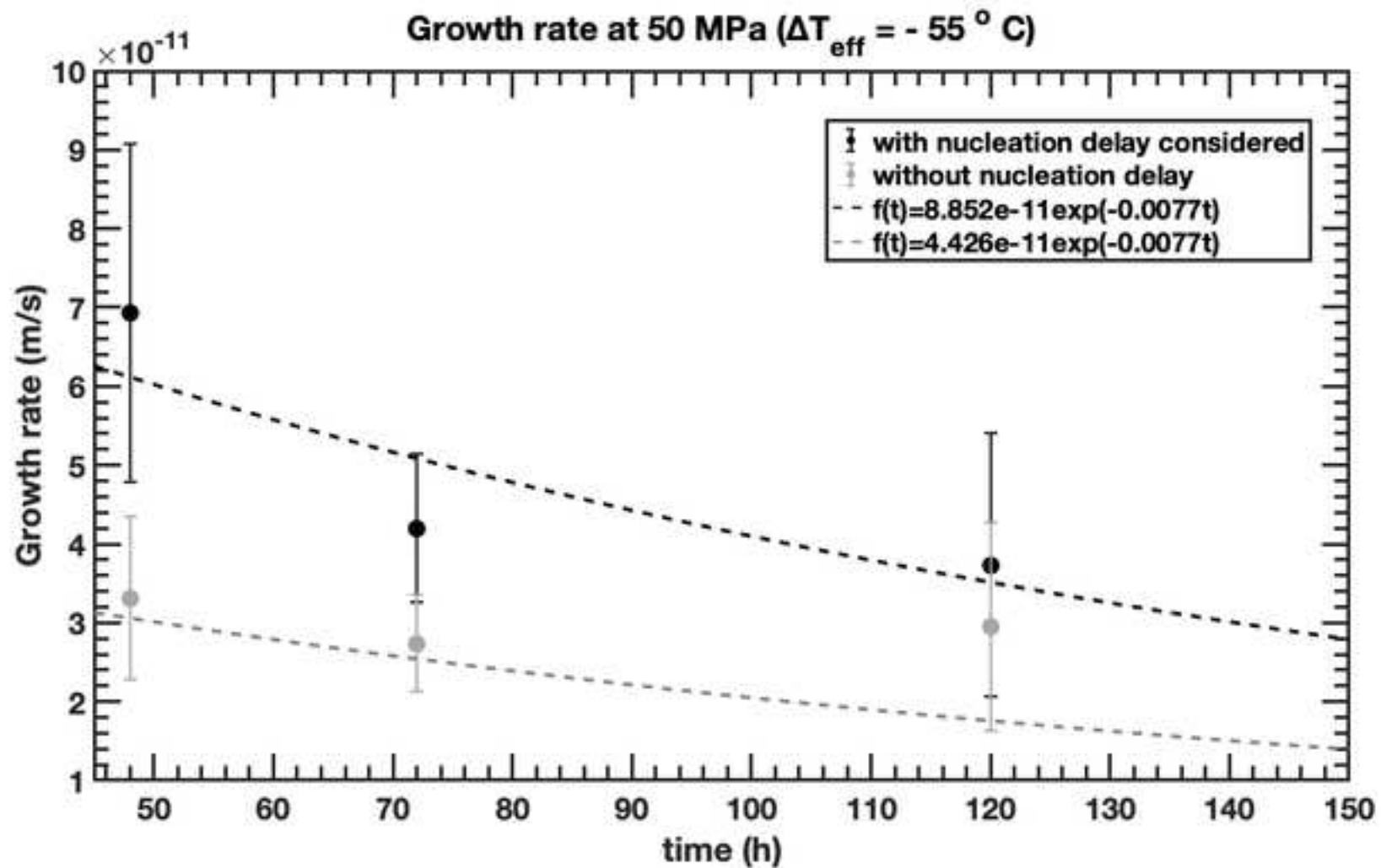


Figure 3







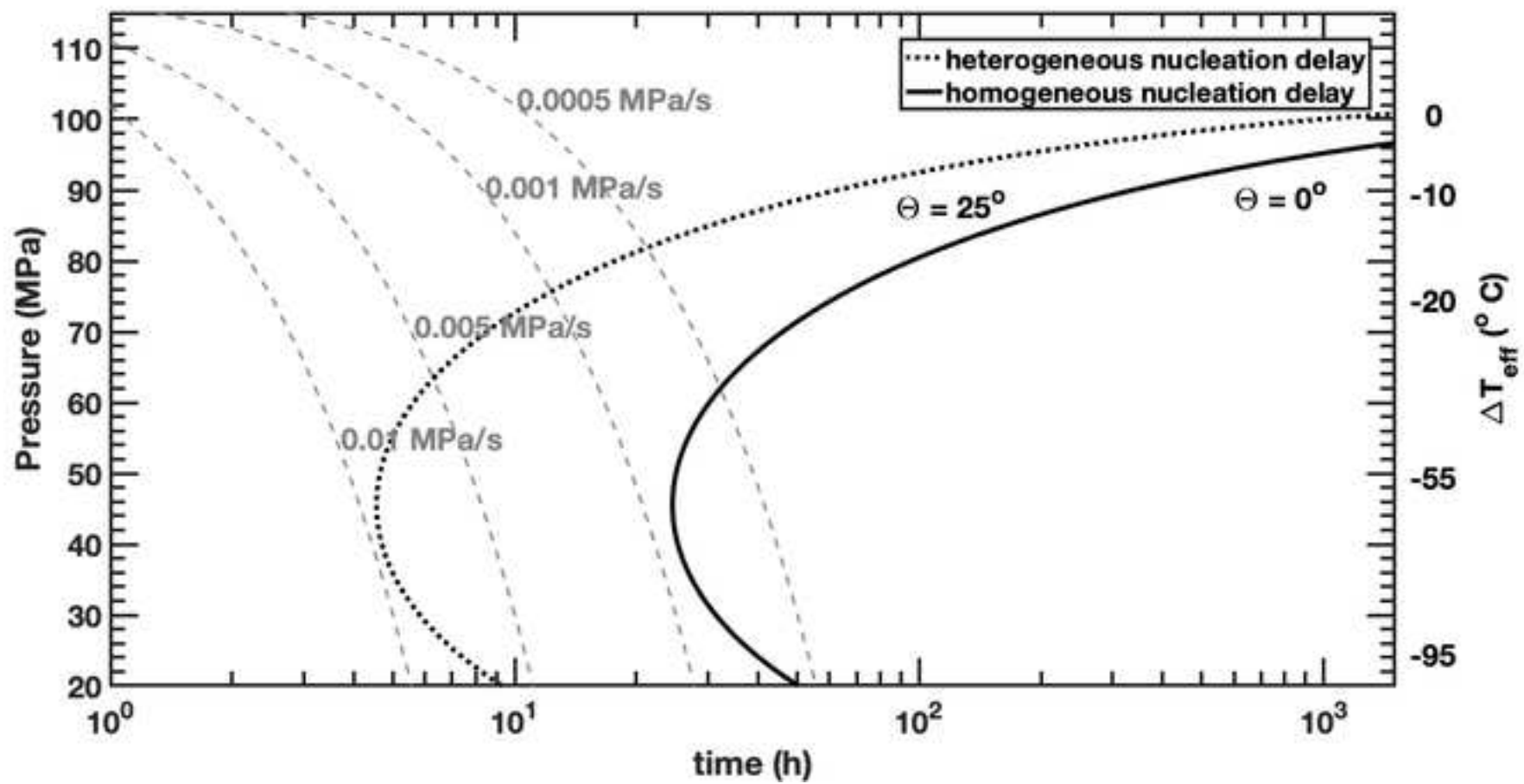
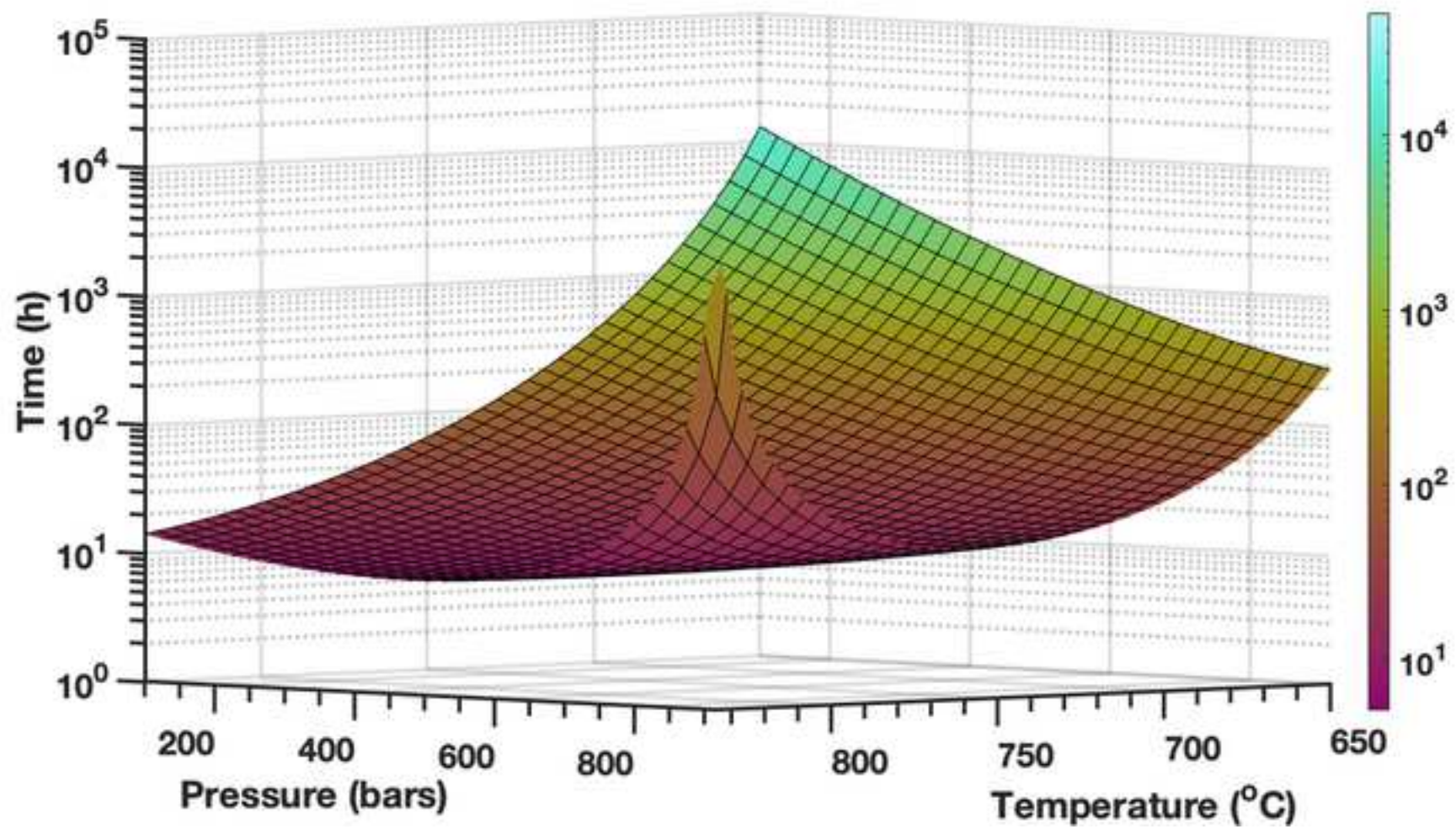


Figure 6



## Sheet1

sample	T (°C)	P <sub>i</sub> (MPa)	P <sub>f</sub> (MPa)	dP/t (MPa/h)	ΔT <sub>eff</sub> (°C)	time at P <sub>f</sub> (h)
LCOD-50	825	200	50	600	-55	0
LCOD-25-4	825	200	25	600	-95	4
LCOD-25-8	825	200	25	600	-95	8
LCOD-25-24	825	200	25	600	-95	24
LCOD-25-51	825	200	25	600	-95	51
LCOD-50-4	825	200	50	600	-55	4
LCOD50-24	825	200	50	600	-55	24
LCOD-50-48	825	200	50	600	-55	48
LCOD-50-72	825	200	50	600	-55	72
LCOD-50-120	825	200	50	600	-55	120
LCOD-75-6	825	200	75	600	-20	6
LCOD-75-30	825	200	75	600	-20	30
LCOD-75-96	825	200	75	600	-20	96
LCOD-75-144	825	200	75	600	-20	144
LCOD-90-72	825	200	90	600	-10	72
LCOD-90-120	825	200	90	600	-10	120
LCOD-90-504	825	200	90	600	-10	504
LCOL200	800	200	200	-	-	168
LCOL200	825	200	200	-	-	168
LCOL200	850	200	200	-	-	168
LCOL200	900	200	200	-	-	168
LCOL150	825	200	200	-	-	168
LCOL50	850	200	200	-	-	168
LCOL50	900	200	200	-	-	168

\* phases: gl – glass; mgt – magnetite; fds – feldspar;

Sheet1

<b>nucleation</b>	<b>phases</b>
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
yes	gl+mgt+fds
yes	gl+mgt+fds
yes	gl+mgt+fds
no	gl+mgt
no	gl+mgt
yes	gl+mgt+fds
yes	gl+mgt+fds
no	gl+mgt
no	gl+mgt
yes	gl+mgt+fds
-	gl+mgt+fds
-	gl+mgt
-	gl+mgt
-	gl+mgt
-	gl+mgt
-	gl+mgt+fds
-	gl+mgt